

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application of: Fan-Nan Lin and Donald H. Macahan

Prior Application: 09/343,376

Examiner: C. Nguyen

Group Art Unit: 1754

For: CATALYTIC REFORMING CATALYST ACTIVATION

**PRELIMINARY AMENDMENT**

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

Pursuant to 37 C.F.R. 1.53(b), Applicant submits the following  
amendments.

**In the Claims**

Please cancel claims 1 - 16 and 18 - 20 without prejudice.

*Clean Copy of Claims* - In compliance with new 37 C.F.R. § 1.121(c),  
please find beginning on the next page clean, amended claim 17. Please substitute  
and enter **this** claim for pending claim 17.

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17. (amended) In a process for reforming naphthene and paraffin-containing petroleum hydrocarbons of gasoline or naphtha boiling range in the presence of molecular hydrogen wherein there is employed a multiple reaction zone system to provide reformates, the improvement which comprises utilizing in said multiple reaction zone system a chlorine-treated catalyst activated by:

(a) reducing a platinum-containing reforming catalyst with hydrogen; and

(b) simultaneously with step (a) contacting said platinum-containing reforming catalyst with a chlorine-containing compound by introducing said chlorine-containing compound serially into each reaction zone of said multiple reaction zone system under conditions to effect decomposition of said chlorine-containing compound thereby providing a chlorine-treated catalyst.

Please add the following new claims:

21. (new) A process according to claim 17 wherein said chlorine-containing compound is selected from the group consisting of tetrachloroethylene, hexachloroethane, carbon tetrachloride, 1-chlorobutane, 1-chloro-2-methyl propane, 2-chloro-2-methyl propane, tertiary butyl chloride, propylene dichloride, perchloroethylene, and mixtures of two or more thereof.

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22. (new) A process according to claim 21 wherein said chlorine-containing compound is perchloroethylene.

23. (new) In a process for reforming naphthene and paraffin-containing petroleum hydrocarbons of gasoline or naphtha boiling range in the presence of molecular hydrogen wherein there is employed a multiple reaction zone system to provide reformates, the improvement which comprises utilizing in said multiple reaction zone system a chlorine-treated catalyst regenerated and activated by:

(a) purging said multiple reaction zone system with an inert gas;

(b) subjecting a deactivated reforming catalyst, contained within said multiple reaction zone system, to an oxidative burning off at a temperature and for a period of time sufficient to remove substantially all carbonaceous deposits thereon thereby providing a substantially carbon free catalyst;

(c) subjecting said substantially carbon free catalyst to an oxygen treatment with a gas containing molecular oxygen at a temperature and for a time sufficient to effect the oxidation of the metals contained in said substantially carbon free catalyst thereby providing an oxidized catalyst;

(d) purging said oxidized catalyst of molecular oxygen thereby providing a purged catalyst;

(e) cooling said purged catalyst thereby providing a cooled catalyst;

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(f) reducing said cooled catalyst with hydrogen, said hydrogen being introduced into a reaction zone of said multiple reaction zone system; and

(g) simultaneously with step (f) contacting said cooled catalyst with a chlorine-containing compound by introducing said chlorine-containing compound serially into each reaction zone of said multiple reaction zone system under conditions to effect decomposition of said chlorine-containing compound thereby providing said chlorine-treated catalyst.

24. (new) A process according to claim 23 wherein said oxidative burning off step (b) is carried out at a temperature in the range of from about 300°F to about 1,300°F and for a period of time in the range of about 4 to about 36 hours.

25. (new) A process according to claim 23 wherein said gas of said oxygen treatment step (c) contains from about 5 to about 15 percent by volume of molecular oxygen.

26. (new) A process according to claim 25 wherein said oxygen treatment step (c) is carried out at a temperature in the range of about 800°F to about 1,150°F.

27. (new) A process according to claim 23 wherein said purged catalyst is cooled to a temperature in the range of about 600°F to about 1,000°F.

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28. (new) A process according to claim 23 wherein step (f) and step (g) are carried out at a temperature in the range of about 500°F to about 1,500°F and at a pressure in the range of from about 0 to about 600 psig.

29. (new) A process according to claim 23 wherein said deactivated reforming catalyst is a platinum-on-alumina reforming catalyst.

30. (new) A process according to claim 29 wherein said platinum-on-alumina reforming catalyst also contains at least one metal selected from the group consisting of rhenium, iridium, ruthenium, tin, palladium, germanium, and combinations of two or more thereof.

31. (new) A process according to claim 23 wherein said nonmetallic chlorine-containing compound is selected from the group consisting of tetrachloroethylene, hexachlorethane, carbon tetrachloride, 1-chlorobutane, 1-chloro-2-methyl propane, 2-chloro-2-methyl propane, tertiary butyl chloride, propylene dichloride, perchloroethylene, and mixtures of two or more thereof.

32. (new) A process according to claim 31 wherein said chlorine-containing compound is perchloroethylene.

#### **In the Specification**

Please add the following new paragraph as the first paragraph on page 1 of the specification:

(new)

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This application is a division of application serial number 09/343,376, filed June 30, 1999, now allowed.

*Clean Copy of Specification Amendments* - In compliance with new 37 C.F.R. §1.121(b), please find beginning on the next page clean, amended paragraphs.

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Please substitute and enter the following amended paragraphs for the current corresponding paragraphs.

**Page 9, Paragraph 1: (amended)**

Generally, the quantity of chlorine-containing compound employed during the chloride treatment must be sufficient to add to the catalyst system from about 0.05 to about 0.3 weight percent chlorine by weight of the total catalyst system (ie. to add about 0.0005 to about 0.003 pounds of chlorine per pound of catalyst), preferably from about 0.1 to about 0.2 weight percent chlorine by weight of the total catalyst system (ie. to add about 0.001 to about 0.002 pounds of chlorine per pound of catalyst). The temperature employed during chloride treatment must be sufficient so as to effect decomposition of the chlorine-containing compound. The chloride treatment can be performed at a temperature of from about 500°F to about 1,500°F, preferably from about 700°F to about 1,200°F, and most preferably from about or 900°F to or about 940°F, and a pressure in the range of about 0 to about 600 psig, preferably about 50 to about 300 psig.

**Page 10, Paragraph 1: (amended)**

containing compound is introduced into the first reactor of the multiple reaction zone system in an amount and for a time to increase to the desired chlorine content, generally to add about 0.05 to about 0.3 weight percent, on the catalyst system of the first reactor. Thereafter, addition of the nonmetallic chlorine-containing

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compound is terminated to the first reactor while hydrogen flow is maintained through the first reactor. Chloride treatment is then commenced to the catalyst contained in the second reactor and continued in the manner as employed in the treatment of the first reactor until the desired chlorine content is established. Thereafter, the catalyst systems of reactor three and reactor four are serially treated in a like manner until the desired chlorine content has been established in each reactor of the series. In carrying out serially the chloride treatment of each reaction zone, hydrogen is flowed continuously through the entire multiple reaction zone system which has been so adapted to permit continuous flow of hydrogen through individual reaction zone.

**Page 19, Paragraph 1: (amended)**

To illustrate the benefits of the activation process of the present invention to the reactor series as employed in Example I, the charged catalyst was activated by reduction with hydrogen at 900°F and then the system was serially treated with perchloroethylene (PCE) to increase the chlorine content by about 0.2 weight percent in each reactor.

**In the Abstract**

*Clean copy of amended Abstract* - Please substitute and enter the following amended Abstract for the current Abstract.

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**Page 28, Paragraph 1: (amended)**

Catalyst activation of a platinum reforming catalyst system contained in a multiple reactor system by simultaneously reducing the catalyst with a sustained hydrogen flow through the multiple reactor system while introducing a nonmetallic chlorine-containing compound serially into each reactor of the multiple reactor system in an amount to add from about 0.05 to about 0.3 weight percent chlorine to the catalyst and thereafter purging the system with about 100 to about 50,000 cubic feet of hydrogen per cubic foot of catalyst prior to commencing use of the treated catalyst system for reforming hydrocarbon feed.

Included herewith is a PTO-Form 1449 listing pertinent pages of a seminar handout titled "Catalytic Reforming Process Technology" by Refining Process Services, Inc..

**Remarks**

This application is submitted in response to the final restriction requirement restricting the prior parent application to multiple inventions. This application is directed to non-elected claims of invention Group III.

In the specification, a cross reference to the prior application has been added and several corrections have been made. In the abstract, the abstract has been amended. In the claims, claims 1 - 16 and 18 - 20 have been cancelled, claim 17 has been amended, and new claims 21 - 32 have been added.

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**Specification Amendments/Corrections**

Attached herewith are copies of pertinent pages from a seminar handout titled "Catalytic Reforming Process Technology" by Refining Process Services, Inc. (RPS), Cheswick PA. In Section 3, on page 2 of the handout, RPS lists compositional data for reforming catalysts including halogen. The chlorine content of such reforming catalysts is listed as being in the range of from 1.0 to 1.2 wt. % chloride. RPS also lists, in section 3, on page 19, UOP R-56 and UOP R-72 as commercial reforming catalysts. UOP R-56 and UOP R-72 are catalysts used in the examples of the instant application. Thus, it appears from the literature that typical commercial reforming catalysts contain in the range of from 1.0 to 1.2 wt. % chloride.

Based on the foregoing paragraph concerning the typical chloride wt. % of typical reforming catalysts, the specification amendments/corrections will be discussed.

On page 10 of the specification, the insertion of the word --- to --- after "increase" on line 2 and the insertion of the word --- add --- after the first occurrence of "to" on line 3 corrects an obvious error. Per the attached RPS document, typical reforming catalysts contain in the range of from 1.0 to 1.2 wt. % chloride. Thus, it is an obvious error in wording to say that contacting the catalyst system with a chlorine-containing compound will "increase the chlorine content

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generally to about 0.05 to about 0.3 wt. %" when the base catalysts, according to RPS, start with 1.0 to 1.2 wt. % chloride. It makes no sense that there would be less chloride on the catalyst after contact with a chlorine-containing compound than there was on the catalyst before contact with such chlorine-containing compound. Thus, it was clearly intended that the contacting of the catalyst system with the chlorine-containing compound would add about 0.05 to about 0.3 wt. % chlorine to the catalyst.

On page 9 of the specification, the replacement of "provide in" with --- add to --- on line 2 and the insertion of --- wt. % chlorine by weight of the total catalyst system (i e. to add about 0.0005 to about 0.0003 --- after "0.3" also corrects an obvious error. It is clear that applicants intended to say on page 9, line 3, that the chlorine-containing compound adds to the catalysts system from about 0.05 to about 0.3 wt. % chlorine by weight of the total catalyst system, rather than about 0.05 to about 0.3 lbs. of chlorine per pound of catalyst, because of the use of the term "wt. %" on page 10, line 3, and in example II on page 19, line 7.

The explanation for the corrections to lines 2 and 3 of page 10 of the specification similarly apply to the correction of line 2 on page 9 of the specification replacing "provide in" with --- add to ---, and applies to the correction of line 6 on page 19 of the specification replacing the second occurrence of "to" with --- by ---, and also applies to the corrections of the abstract of the disclosure on page 28.

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**Claim Amendments**

Support for the amendments to claim 17 can be found at page 8, lines 3 - 7; and page 10, lines 15 - 19; and in original claim 1.

Support for new claims 21 - 22, 24 - 27, and 29 - 32 can be found in original claims 5 - 6, 8 - 11, and 13 - 16.

Support for new claim 23 can be found in original claims 7 and 18, and at page 9, lines 5 - 6.

Support for new claim 28 can be found in the specification at page 9, lines 7 - 10; and in original claim 12.

**Abstract Amendments**

Support for the amendments to the abstract can be found at page 9, lines 1 - 5, as corrected.

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Applicants respectfully request an early notice of allowance.

Respectfully submitted

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Date Of Deposit: July 31, 2001

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July 31, 2001  
(Date)

Jeffrey R. Anderson  
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***VERSION WITH MARKINGS TO SHOW CHANGES IN CLAIMS***

17. (amended) In a process for reforming naphthene and paraffin-containing petroleum hydrocarbons of gasoline or naphtha boiling range in the presence of molecular hydrogen wherein there is employed [in series a plurality of catalytic reaction zones] a multiple reaction zone system to provide reformates, the improvement which comprises utilizing in said [catalytic reaction zones] multiple reaction zone system a chlorine-treated catalyst activated by [the process of claim 1.];

(a) reducing a platinum-containing reforming catalyst with hydrogen;  
and

(b) simultaneously with step (a) contacting said platinum-containing reforming catalyst with a chlorine-containing compound by introducing said chlorine-containing compound serially into each reaction zone of said multiple reaction zone system under conditions to effect decomposition of said chlorine-containing compound thereby providing a chlorine-treated catalyst.

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***VERSION WITH MARKINGS TO SHOW CHANGES IN SPECIFICATION*****Page 9, Paragraph 1: (amended)**

Generally, the quantity of chlorine-containing compound employed during the chloride treatment must be sufficient to [provide in] add to the catalyst system from about 0.05 to about 0.3 weight percent chlorine by weight of the total catalyst system (ie. to add about 0.0005 to about 0.003 pounds of chlorine per pound of catalyst), preferably from about 0.1 to about 0.2 weight percent chlorine by weight of the total catalyst system (ie. to add about 0.001 to about 0.002 pounds of chlorine per pound of catalyst). The temperature employed during chloride treatment must be sufficient so as to effect decomposition of the chlorine-containing compound. The chloride treatment can be performed at a temperature of from about 500°F to about 1,500°F, preferably from about 700°F to about 1,200°F, and most preferably from about or 900°F to or about 940°F, and a pressure in the range of about 0 to about 600 psig, preferably about 50 to about 300 psig.

**Page 10, Paragraph 1: (amended)**

containing compound is introduced into the first reactor of the multiple reaction zone system in an amount and for a time to increase to the desired chlorine content, generally to add about 0.05 to about 0.3 weight percent, on the catalyst system of the first reactor. Thereafter, addition of the nonmetallic chlorine-containing compound is terminated to the first reactor while hydrogen flow is maintained through the first reactor. Chloride treatment is then commenced to the catalyst

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contained in the second reactor and continued in the manner as employed in the treatment of the first reactor until the desired chlorine content is established. Thereafter, the catalyst systems of reactor three and reactor four are serially treated in a like manner until the desired chlorine content has been established in each reactor of the series. In carrying out serially the chloride treatment of each reaction zone, hydrogen is flowed continuously through the entire multiple reaction zone system which has been so adapted to permit continuous flow of hydrogen through individual reaction zone.

**Page 19, Paragraph 1: (amended)**

To illustrate the benefits of the activation process of the present invention to the reactor series as employed in Example I, the charged catalyst was activated by reduction with hydrogen at 900°F and then the system was serially treated with perchloroethylene (PCE) to increase the chlorine content [to] by about 0.2 weight percent in each reactor.

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*VERSION WITH MARKINGS TO SHOW CHANGES IN ABSTRACT***Page 28, Paragraph 1: (amended)**

Catalyst activation of a platinum reforming catalyst system contained in a multiple reactor system by simultaneously reducing the catalyst with a sustained hydrogen flow through the multiple reactor system while introducing a nonmetallic chlorine-containing compound serially into each reactor of the multiple reactor system in an amount to [provide] add from about 0.05 to about 0.3 weight percent chlorine [on] to the catalyst and thereafter purging the system with about 100 to about 50,000 cubic feet of hydrogen per cubic foot of catalyst prior to commencing use of the treated catalyst system for reforming hydrocarbon feed.

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